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MICROSCOPIC SCALE SIMULATIONS OF SODA-LIME-SILICA USING MOLECULAR DYNAMICS

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Abstract. Present work is focused on the investigation of soda-lime-silica glass. The material is completely amorphous, therefore a 3D random network generation was done to produce the initial geometry. The systems were verified with neutron diffraction results. The simulation was done in two steps. First the geometry was generated and verified, then the system was submitted to simple mechanical loading such as unidirectional compression. The overall goal of the simulation is to compare the results of the discrete mechanics behavior with macroscopically measurable continuum mechanics parameters as the elasticity.

1 INTRODUCTION

Thanks to its transparency structural glass elements has become one of the most popular architectural material nowadays. The primary component of these structural elements made of soda-lime-silica. The material itself has a significantly high strength, but most of the design standards make serious restrictions about the effective tensile strength. Thanks to its amorphous atomic structure and covalent bounds soda-lime-silica has a highly brittle macroscopic fracture. Several works were done to investigate the atomic structure of pure silica [1]-[4], calcium silicate [5] and soda-lime-silica [6], although relatively few referred to the micro-mechanical behavior of the material.

A mesoscopic investigation was carried out to determine the macroscopic stress increments on a structural glass plate [7]-[8]. Scratches and flaws were measured in a nanometer scale. Finite element simulations were used to calculate stress distribution on the surfaces submitted

to simple mechanical deformations (shear and tension). It was not clear that a continuum mechanical approach can be used in such small scale. Present paper is devoted to define a representative volume element (RVE) using molecular dynamics (MD) simulation for the soda-lime-silica, which contains all the relevant mechanical properties of the material. During the simulation, several atomic structures were generated with different sizes. The smallest structure contained 71, the largest 71320 atoms, the size of the cube edge was respectively 10 to 100 Å.

3 METHOD

3.1 Molecular dynamics

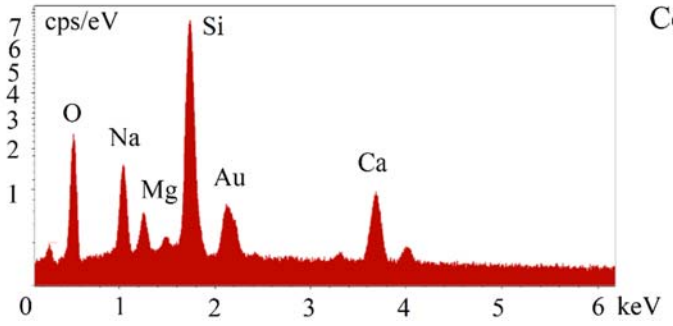
Molecular dynamics is a numerical method which calculates the physical movements of the atoms. The method basically solves the second Newtonian law in a period of time using an explicit numerical integration. The interaction between the atoms is defined using interatomic potentials, where the interacting forces are calculated by the derivate of the potential functions.

The simulated atomic systems had several properties which could be compared to real life experimental results, such as structure factor or angle distribution. These parameters were defined previously by neutron diffractography [9].

The temperature was controlled with a Nosé–Hoover thermostat. The simulations were performed with LAMMPS.

3.2 Composition

The exact composition of the material was determined using energy-dispersive X-ray spectroscopy (EDX).



Composition

Chemical element	Mass ration
Silicon	33.27%
Oxygen	43.62%
Sodium	9.93%
Calcium	10.68%
Magnesium	2.50%

Figure 1: Composition of the investigated material

In Fig. 1 a large amount of gold can be recognized. The presence of gold can be explained by the preparation method for the scanning electron microscopy. All the specimens were coated with a thin gold layer to prevent electrostatic charge. The amount of the magnesium as a contamination was very low compared to the base materials. Therefore from the first group of the simulations the gold and the magnesium were excluded. The composition of the material was considered 34.12 % silicon, 44.74 % oxygen, 10.19 % sodium and 10.96 % calcium.

3.3 Interatomic potentials

We have used the potential model developed for silica by Van Beest, Kramer, and Van Santen. The BKS potential was expanded with the classic Coulombic interaction. The potential function can be written as follows:

$$\Phi(r_{ij})=\frac{q_i q_j e^2}{r_{ij}}+A_{ij}\exp\left(-\frac{r_{ij}}{\rho_{ij}}\right)-\frac{C_{ij}}{r_{ij}^6}, \tag{1}$$

where q_i and q_j are the charge of the atoms, r_{ij} is the distance between atoms, e is the dielectric constant. A_{ij}, ρ_{ij} and C_{ij} are the empirical parameters of the BKS potential. The numerical values of the parameters are shown in Tab. 1. The cutoff distance was set to 10 Å.

Table 1: Parameters of BKS potential [6]

Pairs	A_{ij} [eV]	ρ_i [Å]	C_{ij} [eV×Å^6]
Si – O	18003.76	0.2052	133.5381
O – O	1388.773	0.36232	175
Na – O	34000	0.1875	10
Na – Na	9500	0.23	0
Ca – O	131400	0.1875	60
Ca – Ca	10000	0.23	0

Atoms	Atomic charges [e]
Silicon	+2.4
Oxygen	-1.2
Sodium	+0.6
Calcium	+1.2

3.4 Initial geometry

The simulations had two steps: first the initial position of the atoms was generated and the system was verified, then the mechanical load was applied.

The simulations were carried out for different bulk sizes. The systems were cubic boxes containing 71, 571, 1926, 4564, 8915, 11688, 15405 and 36516 atoms, where the cube edge sizes are respectively 10, 20, 30, 40, 50, 55, 60, and 80 Å (Fig. 2). The density of the system was assumed $\rho = 2.503\ g/cm^3$.

The atoms were randomly placed in the simulation area with defined rate. The deposition frequency of the silicon was 3.8 atom/ps, for oxygen 8.7 atom/ps, for sodium 1.4 atom/ps and for calcium 0.85 atom/ps. The construction of the system was carried out on constant 300 K. No heating and quenching was necessary, because the particles had the right amount of time to reach their proper position in the structure with the deposition rates mentioned above.

For the explicit numerical integrator a time step of $\Delta t = 10^{-4}\ ps$ was used. During the generation process periodic boundaries were assumed in all three directions.

The deposition was carried out in the first 80 % of the generation, then the system was equilibrated. For example during the creation of 50 Å box the atoms were deposited in 600 ps, then 150 ps was used to equilibrate the system.

During the simulations NVT (constant number of atoms, volume and temperature) technique was applied.

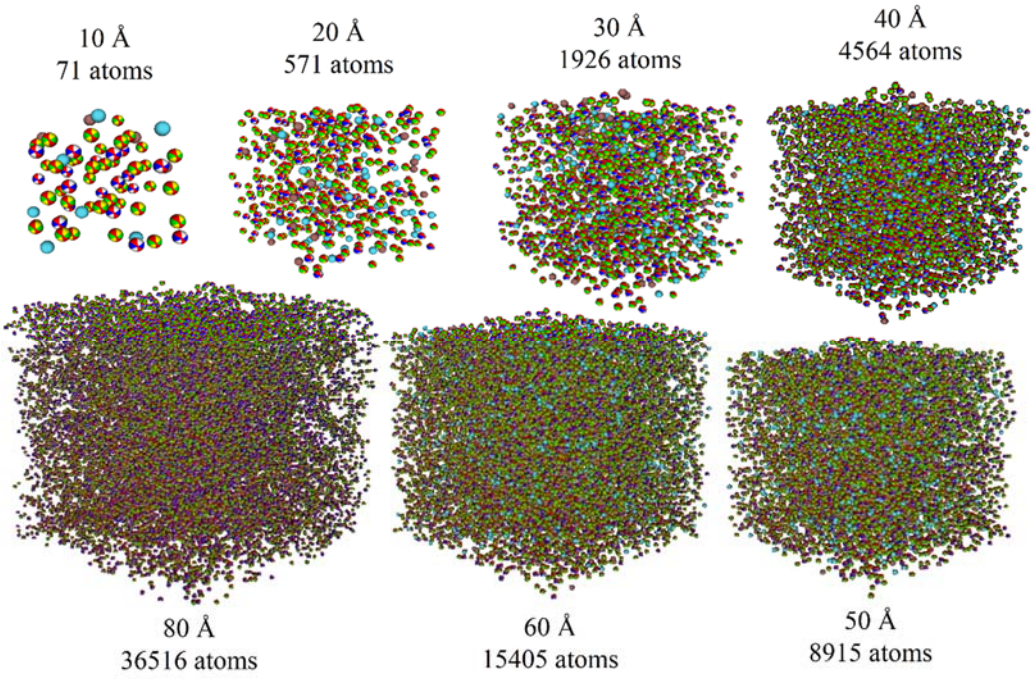


Figure 2: Different size atomic structures

4 VERIFICATION

The verification procedure shown below is carried out on a 60 Å size system. Thanks to that the structure factor is not a sensitive parameter not only the complete function is going to be shown, but the partial pair distribution functions too.

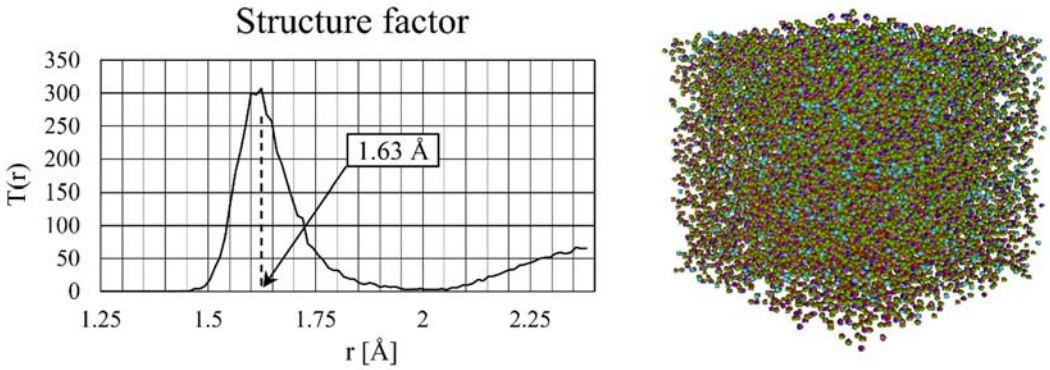


Figure 3: Structure factor of 60 Å size system

The structure factor shown in Fig. 3 is the histogram of the calculated distances between all the atoms. The largest peak is calculated at 1.63 Å, which corresponds to the intra-tetrahedral Si–O distances. According to [9] the same distance value is present in the soda-lime-silica used for structural purposes.

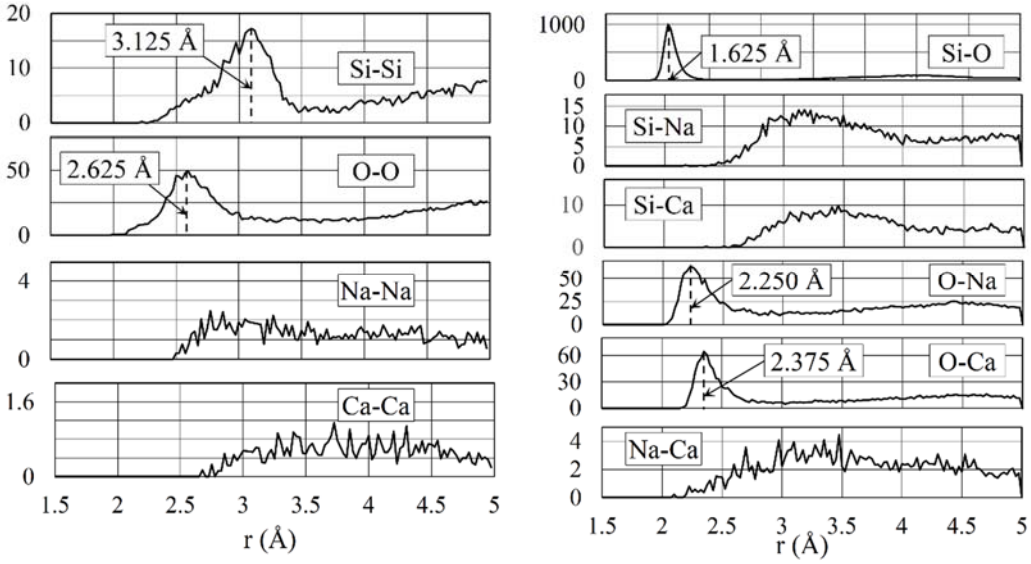


Figure 4: Partial pair distribution functions

Fig. 4 shows the partial pair distribution functions of the system. On the Si-Si diagram the first and largest peak appears at 3.125 Å, the distance is a bit smaller considering O-O pairs: 2.625 Å. Na-Na and Ca-Ca curves are not consistent, although are very similar compared to the measured ones in [9].

For different atoms the first and largest peak is at 1.625 Å which corresponds to the Si-O pairs which are the most common is the system. This peak can be recognized on the structure factor as well. Among the relevant peaks O-Na pairs has a significant value at 2.250 Å, and O-Ca pairs has a peak at 2.375 Å. Si-Na, Si-Ca and Na-Ca functions does not carry any evaluable results, although the shapes are also consistent with the curves measured with neutron diffractography. In Tab. 2 the comparison could be seen between the measured [9] and the simulated values.

Table 2: Distance peak comparison between simmulated and measured [9] values

Atomic pairs	Simulated distance [Å]	Measured with neutron diffraction [Å]
Si-Si	3.125	3.12-3.19
O-O	2.625	2.67
Na-Na	N/A	N/A
Ca-Ca	N/A	N/A
Si-O	1.625	1.629±0.005
Si-Na	3.200	N/A
Si-Ca	3.475	N/A
O-Na	2.250	2.25
O-Ca	2.375	2.35
Na-Ca	N/A	N/A

5 UNIDIRECTIONAL COMPRESSION

After the generation of various atomic structures unidirectional compression was applied. The simulations were carried out on constant temperature of 300 K with NVT method. The load was applied with the strain rate of 10^9 1/s with a loading plane. The plane has an effect on every atom with a user defined function. The force is calculated in the function of the distance between the plane and the atoms.

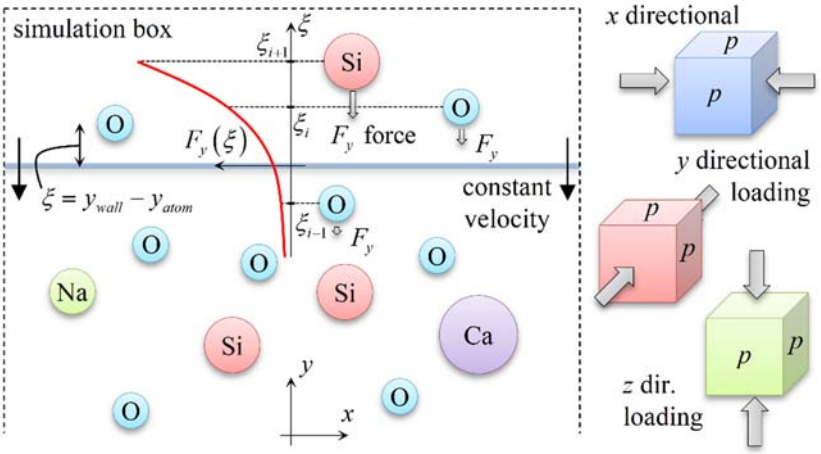


Figure 5: Working scheme of loading

In Fig. 5 ξ is the distance between the wall and the atom. $F_y(\xi)$ is the force applied on the atoms. Stress is calculated by summarizing the forces applied by the wall and then divided by the area where it was applied. This way we could simply calculate a parameter used in continuum mechanics from discrete results. The movement of the wall was controlled with constant velocity.

The load was applied always in one direction, along the other two axes the boundaries were considered periodically. Therefore the simulation was developed to model a thin layer of soda-lime-silicate.

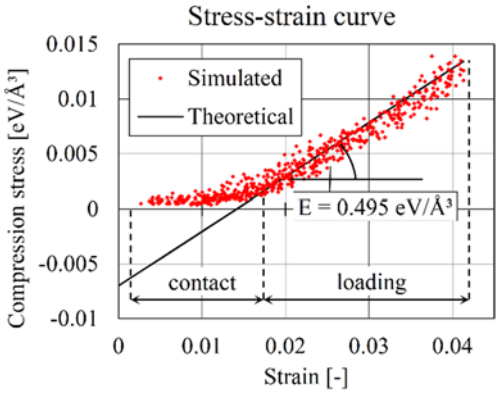


Figure 6: Stress-strain curve of uniaxial compression on a 55 Å size system

In Fig. 6 two stages can be recognized. During the first – called contact – stage the wall touches the outer layer of the particles and produces its effects. In the second stage a constant linear compression could be seen with the elastic moduli of glass calculated for present boundary conditions. In the figure a visible scattering could be recognized which is the effect of the thermal vibrations.

6 FUTURE PLANS

We have created and verified atomic structures of amorphous soda-lime-silica virtually. On the verified model we applied basic unidirectional compression to calculate continuum mechanics parameters. We have found that the simulated Young's modulus has a good correlation with the macroscopically calculated one, although a slight scattering was observed due to the thermal vibration. Furthermore we would like to know in which size the deviation of the elastic moduli can be neglected, this way we could define the size of the macroscopic representative volume element (RVE).

In the future we are planning to calculate the effect of the loading on the pair distribution functions as on the whole structure factor.

Our intention is to apply shear load also, and investigate the effect of the deviatoric shear strain on the amorphous atomic structure of soda-lime-silica.

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